Longitudinal spin decoherence in spin diffusion in semiconductors

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We set up a set of many-body kinetic Bloch equations with spacial inhomogeneity. We reexamine the widely adopted quasi-independent electron model and show the inadequacy of this model in studying the spin transport. We further point out a new decoherence effect based on interference along the direction of diffusion in spin transport due to the so called inhomogeneous broadening effect in the Bloch equations. We show that this inhomogeneous broadening can cause decoherence alone, even in the absence of the scattering and that the resulting decoherence is more important than the dephasing effect due to both the D'yakonov-Perel' (DP) term and the scattering.

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Study of spintronics has attracted tremendous attention in recent years, both in theoretical and experimental circles,[1] thanks to the discovery of the long-lived (sometimes > 100 ns) coherent electron spin states in n-typed semiconductors.[2,3,4,5,6,7] Possible applications of spintronics include qubits for quantum computers, quantum memory devices, spin transistors, and spin valves etc. The last two applications involve transporting spin polarized electrons from a place to another by means of an electrical or diffusive current. Therefore, it is of great importance to study the spin transport. Apart from the great number of works on spin injection, there are only a few experimental reports on coherent spin transport over macroscopic distance.[3,8,9] On theoretical aspect, most works are based on a quasi-independent electron model and focused on the diffusive transport regime,[10,11,12,13,14,15] where equations for spin polarized currents can be set up and the longitudinal spin dephasing, generally referred to as spin diffusion length can be achieved. In these theories, the mechanism for the spin relaxation is assumed to be due to the spin-flip scattering. In the absence of the scattering, the spin polarization will not decay in a nonmagnetic sample. In Ref. 16, Takahashi *et al.* calculated the scattering induced spin relaxation time associated with the spin diffusion starting from the many body kinetic equations.

Of particular interest to the spin transport theory in semiconductors has been the question as to whether the quasi-independent electron model can adequately account for the experimental results or whether many-body processes are important. Flatte *et al.* have concluded that an independent electron approach is quite capable of explaining measurements of spin lifetimes in the diffusive regime.[17] In this paper, we reexamine this issue from a full many-body transport theory and show the inadequacy of the independent electron model in describing the spin transport. We also propose a mechanism that may cause strong longitudinal spin decoherence in addition to the spin dephasing due to scattering. The new mechanism is based on the interference effect due to the wavevector dependence of the spin densities along the spacial gradients in the spin diffusion. This wavevector dependence can be considered as some sort of "inhomogeneous broadening", which can cause spin decay alone, even in the absence of scattering.

Recently, we have presented a many-body kinetic theory to describe the spin precession and dephasing in insulating samples as well as n-doped samples.[18,19,20] In this paper we extend this theory to the spacial inhomogeneous regime and obtain the many-body transport equations necessary to investigate the spin diffusion in n-doped GaAs. Here, we only focus on the spin transport inside the semiconductor and avoid the problem of spin injection at the boundary. Based on the two-spin-band model[19] in the conduction bands, we construct the semiconductor Bloch equations by using the nonequilibrium Green function method with gradient expansion as well as the generalized Kadanoff-Baym Ansatz[21] as follows:

$$\frac{\partial \rho(\mathbf{R}, \mathbf{k}, t)}{\partial t} - \frac{1}{2}\big\{\nabla_{\mathbf{R}}\bar{\varepsilon}(\mathbf{R}, \mathbf{k}, t), \nabla_{\mathbf{k}}\rho(\mathbf{R}, \mathbf{k}, t)\big\} + \frac{1}{2}\big\{\nabla_{\mathbf{k}}\bar{\varepsilon}(\mathbf{R}, \mathbf{k}, t), \nabla_{\mathbf{R}}\rho(\mathbf{R}, \mathbf{k}, t)\big\} - \frac{\partial \rho(\mathbf{R}, \mathbf{k}, t)}{\partial t}\bigg|_c = \frac{\partial \rho(\mathbf{R}, \mathbf{k}, t)}{\partial t}\bigg|_s . \quad (1)$$

Here $\rho(\mathbf{R}, \mathbf{k}, t)$ represents a single particle density matrix. The diagonal elements describe the electron distribution functions $\rho_{\sigma\sigma}(\mathbf{R}, \mathbf{k}, t) = f_\sigma(\mathbf{R}, \mathbf{k}, t)$ of wave vector $\mathbf{k}$ and spin $\sigma(= \pm 1/2)$ at position $\mathbf{R}$ and time t. The off-diagonal elements $\rho_{\sigma-\sigma}(\mathbf{R}, \mathbf{k}, t)$ describe the inter-spin-band polarization components (coherences) for the spin coherence. The quasi-particle energy $\bar{\varepsilon}_{\sigma\sigma'}(\mathbf{R}, \mathbf{k}, t)$, in the presence of a moderate magnetic field $\mathbf{B}$ and with the DP mechanism[22] included, can be written as

$$\bar{\varepsilon}_{\sigma\sigma'}(\mathbf{R}, \mathbf{k}, t) = \varepsilon_k \delta_{\sigma\sigma'} + \big[g\mu_B\mathbf{B} + \mathbf{h}(\mathbf{k})\big] \cdot \frac{\vec{\sigma}_{\sigma\sigma'}}{2}$$
$$- e\psi(\mathbf{R}, t) + \Sigma_{\sigma\sigma'}(\mathbf{R}, \mathbf{k}, t) . \quad (2)$$

Here $\varepsilon_k = k^2/2m^*$ is the energy spectrum with m^* denoting electron effective mass, $-e$ is the electron charge and $\vec{\sigma}$ are the Pauli matrices and $\mathbf{h}(\mathbf{k})$ originate from the DP mechanism which contains both the Dresselhaus[23] and the Rashba terms.[24] In this paper, we only consider the first one. For [001] quantum well, it can be written as[25] $h_x(\mathbf{k}) = \gamma k_x(k_y^2 - \kappa_z^2)$, $h_y(\mathbf{k}) = \gamma k_y(\kappa_z^2 - k_x^2)$, with κ_z^2 denoting the average of the operator $-(\partial/\partial z)^2$ over the electronic state of the lowest subband. $\gamma = (4/3)(m^*/m_{cv})(1/\sqrt{2m^{*3}E_g})(\eta/\sqrt{1 - \eta/3})$ and $\eta = \Delta/(E_g + \Delta)$. Here E_g denotes the band gap, Δ represents the spin-orbit splitting of the valence band, and m_{cv} is a constant close in magnitude to the free electron mass m_0.[26] The electric potential $\psi(\mathbf{R}, t)$ satisfies the Poisson equation

$$\nabla_{\mathbf{R}}^2 \psi(\mathbf{R}, t) = -e[n(\mathbf{R}, t) - n_0(\mathbf{R})]/\epsilon , \qquad (3)$$

where $n(\mathbf{R}, t) = \sum_{\sigma \mathbf{k}} f_\sigma(\mathbf{R}, \mathbf{k}, t)$ is the electron density at position $\mathbf{R}$ and time t, and $n_0(\mathbf{R})$ is the background positive electric charge density. $\Sigma_{\sigma\sigma'}(\mathbf{R}, \mathbf{k}, t) = -\sum_{\mathbf{q}} V_{\mathbf{q}} \rho_{\sigma\sigma'}(\mathbf{R}, \mathbf{k} - \mathbf{q}, t)$ is the Hartree-Fock self-energy, with $V_{\mathbf{q}}$ denoting the Coulomb matrix element. In 2D case, $V_{\mathbf{q}}$ is given by

$$V_{\mathbf{q}} = \sum_{q_z} \frac{4\pi e^2}{\epsilon_0(\mathbf{q}^2 + q_z^2 + \kappa^2)} |I(iq_z)|^2, \qquad (4)$$

in which $\kappa = 2e^2 m^*/\epsilon_0 \sum_\sigma f_\sigma(K = 0)$ is the inverse screening length, with ε_0 being the static dielectric constant. The form factor $|I(iq_z)|^2 = \pi^2 \sin^2 y/[y^2(y^2 - \pi^2)^2]$ with $y = q_z a/2$. It is noted that when one takes only the diagonal elements $\rho_{\sigma\sigma}$ of Eq. (1) and neglects all off-diagonal ones $\rho_{\sigma-\sigma}$, the first three terms on the left hand side of the equation correspond to the drift terms in the classical Boltzmann equation, modified with the DP terms and self energy from the Coulomb Hartree term. $\frac{\partial \rho(\mathbf{R}, \mathbf{k}, t)}{\partial t}|_c$ and $\frac{\partial \rho(\mathbf{R}, \mathbf{k}, t)}{\partial t}|_s$ in the Bloch equations (1) are the coherent and scattering terms respectively, with the symbols $|_c$ and $|_s$ standing for "coherent" and "scattering". The components of the coherent terms can be written as[21,20]

$$\left.\frac{\partial f_\sigma}{\partial t}\right|_c = -2\text{Im}[\bar{\varepsilon}_{\sigma-\sigma}\rho_{-\sigma\sigma}], \qquad (5)$$

$$\left.\frac{\partial \rho_{\sigma-\sigma}}{\partial t}\right|_c = i[\bar{\varepsilon}_{\sigma\sigma} - \bar{\varepsilon}_{-\sigma-\sigma}]\rho_{\sigma-\sigma} + i\bar{\varepsilon}_{\sigma-\sigma}[f_{-\sigma} - f_\sigma] . \qquad (6)$$

While the scattering terms $\left.\frac{\partial \rho(\mathbf{R}, \mathbf{k}, t)}{\partial t}\right|_s$ are given in detail in Eqs. (5) and (7) of Ref. 20.

The Bloch equations (1) can be reduced to their counterpart in the independent electron approach as follows. The DP term forms an effective magnetic field. It can flip the spin-up electrons to the spin-down ones, and *vice versa*. The DP term combines with the scattering will result in a longitudinal spin dephasing.[22,19,20] By applying the relaxation time approximation to describe this dephasing and discarding the spin coherences $\rho_{\sigma-\sigma}(\mathbf{R}, \mathbf{k}, t)$

as well as the DP term (to avoid double counting) and carrying out the summation over $\mathbf{k}$, one obtains the the continuity equation for electrons of spin σ

$$\frac{\partial n_\sigma(\mathbf{R}, t)}{\partial t} - \frac{1}{e}\nabla_{\mathbf{R}}\cdot\mathbf{J}_\sigma(\mathbf{R}, t) = -\frac{n_\sigma(\mathbf{R}, t) - n_0(\mathbf{R}, t)}{\tau_s} , \qquad (7)$$

in which $n_0(\mathbf{R}, t) = [n_\sigma(\mathbf{R}, t) + n_{-\sigma}(\mathbf{R}, t)]/2$ is the total electron number at $\mathbf{R}$. $\mathbf{J}_\sigma(\mathbf{R}, t) = \sum_{\mathbf{k}}(-e)\mathbf{v}_{\sigma\mathbf{k}} f_\sigma(\mathbf{R}, \mathbf{k}, t)$ is the electric current of spin σ. The spin dependent velocity is $\mathbf{v}_{\sigma\mathbf{k}} = \nabla_{\mathbf{k}}\bar{\varepsilon}_{\sigma\sigma}(\mathbf{R}, \mathbf{k}, t)$ where $\bar{\varepsilon}_{\sigma\sigma}(\mathbf{R}, \mathbf{k}, t)$ is given by Eq. (2) but without the DP term $\mathbf{h}(\mathbf{k})$. By applying the relaxation time approximation to describe the momentum scattering and keeping terms of the the lowest order (*ie.*, neglecting terms containing $\rho_{\sigma-\sigma}$) and carrying out the summation over $\mathbf{k}$, one obtains the expression for the current in the steady state:

$$\mathbf{J}_\sigma(\mathbf{R}, t) = n_\sigma(\mathbf{R}, t)e\mu\mathbf{E}(\mathbf{R}, t) + eD\nabla_{\mathbf{R}}n_\sigma(\mathbf{R}, t) . \qquad (8)$$

Here μ and D represent the electron mobility and diffusion constant respectively. Equations (7) and (8) are the diffusion equations in the independent electron approach.[10,11,13,14,15]

One can see from the derivation of above diffusion equations that, by summing over $\mathbf{k}$, the $\mathbf{k}$ dependence of the coefficients of $\nabla_{\mathbf{R}}\rho(\mathbf{R}, \mathbf{k}, t)$ in the Bloch equation (1) is removed. This will not cause any problem when there is no spin precession. However, when the electron spin precesses along with the diffusion, *e.g.* in the presence of a magnetic field or of an effective one (*i.e.* the DP term), this kind of $\mathbf{k}$ dependence may cause additional decoherence.

To reveal this effect , we apply the above kinetic equation to study the stationary state in the plane of an n-doped GaAs quantum well (QW), with its growth direction along the z-axis. The width of the QW is assumed to be small enough so that only the lowest subband is important. We assume one side of the sample ($x = 0$) is connected with an Ohmic contact which gives constant spin polarized injection. In this study, we assume the electric field $E = 0$. The diffusion is along the x direction. The electron distribution functions at the interface are assumed to be the Fermi distributions

$$f_\sigma(0, \mathbf{k}, t) \equiv f_\sigma^0(\mathbf{k}) = \{\exp[(\varepsilon_k - \mu_\sigma)/T] + 1\}^{-1} , \qquad (9)$$

with T being the temperature and μ_σ representing the electron chemical potential of spin σ. The spin coherence at the interface is assumed to be zero

$$\rho_{\sigma-\sigma}(0, \mathbf{k}, t) \equiv 0 . \qquad (10)$$

It is understood that the boundary condition here is an approximation to describe the distributions just after the injection of the *spin polarization* from the Ohmic contact. There is no net charge injection into the QW and

the well is kept charge neutral everywhere. Actually, this boundary condition does not necessarily come from the injection at the interface. It can also be produced in the center of semiconductors by a circularly polarized cw laser.

We first consider a much simplified case by neglecting the DP terms $\mathbf{h}(\mathbf{k})$, the self energies as well as the scattering terms in the Bloch equations (1). The simplified equations are therefore as follows

$$\frac{k_x}{m^*}\partial_x f_\sigma(x,\mathbf{k}) - g\mu_B B\mathrm{Im}\big[\rho_{-\sigma,\sigma}(x,\mathbf{k})\big] = 0, \qquad (11)$$

$$\frac{k_x}{m^*}\partial_x \rho_{\sigma-\sigma}(x,\mathbf{k}) - i\frac{g\mu_B B}{2}\Delta f_\sigma(x,\mathbf{k}) = 0. \qquad (12)$$

Here we take the magnetic field $\mathbf{B}$ along the x-axis. $\Delta f_\sigma(x,\mathbf{k}) = f_\sigma(x,\mathbf{k}) - f_{-\sigma}(x,\mathbf{k})$. The solution for this simplified equations with the boundary conditions (9) and (10) can be written out directly

$$\Delta f_\sigma(x,\mathbf{k}) = \Delta f^0(\mathbf{k})\cos\frac{g\mu_B Bm^* x}{k_x}, \qquad (13)$$

$$\rho_{\sigma-\sigma}(x,\mathbf{k}) = \frac{i}{2}\Delta f^0(\mathbf{k})\sin\frac{g\mu_B Bm^* x}{k_x}. \qquad (14)$$

Equations (13) and (14) clearly show the effect of the k-dependence to the spin precession along the diffusion direction. For each fixed k_x, the spin precesses along the diffusion direction with fixed period without any decay. Nevertheless, for different k_x the period is different. The total difference of the electron densities with different spin is the summation over all wavenumbers $\Delta N = \sum_{\mathbf{k}}\Delta f_\sigma(x,\mathbf{k})$. It is noted that the phase at the contact $x = 0$ for different k_x is all the same. However, the speed of the phase of spin precession is different

for different k_x. Consequently, when x is large enough, spins with different phases may cancel each other. This can further be seen from Fig. 1 where the electron densities $N_\sigma = \sum_{\mathbf{k}} f_\sigma(x,\mathbf{k})$ for up and down spin are plotted as functions of position x. The boundary electron densities at $x = 0$ are $N_{1/2}(0) = 2.05 \times 10^{11}$ cm^{-2} and $N_{-1/2}(0) = 1.95 \times 10^{11}$ cm^{-2}. We take $B = 1$ T and $T = 200$ K. In order to show the transverse spin dephasing, we plot in the same figure the incoherently summed spin coherence $\rho(t) = \sum_{\mathbf{k}} |\rho_{\frac{1}{2}-\frac{1}{2}}(x,\mathbf{k})|$. It is understood that both the true dissipation and the interference among the $\mathbf{k}$ states may contribute to the decay. The decay due to interference is caused by the different precessing rates of electrons with different wavevectors. For finite system, this leads to reversible loss of coherence among electrons.[27,28] We refer to this kind of loss of coherence as decoherence. Whereas for the true dissipation, the coherence of the electrons is lost irreversibly.[29,27,28] The irreversible loss of coherence is termed dephasing in this paper. The incoherent summation is therefore used to isolate the irreversible decay from the decay caused by interference.[19,29] From the figure, one can see clearly the longitudinal decoherence caused by the interference effect. It is also noted from the figure that ρ does not decay with the distance. This is consistent with the fact that there is no scattering in Eqs. (13) and (14) and the decay comes only from the interference effect.

Facilitated with the above understanding, we turn to the spin diffusion problem with the DP terms, self-energies and scattering included. We take $\mathbf{B} = \mathbf{E} = 0$. By substituting the quasi-particle energy $\varepsilon_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t)$ [Eq. (2)] into the Bloch equations (1), the first three terms in Eqs. (1) can be written as

$$\partial_t\rho_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t) + e\partial_x\psi(\mathbf{R},t)\partial_{k_x}\rho_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t) - \frac{1}{2}\sum_{\sigma_1}\Big[\partial_x\Sigma_{\sigma\sigma_1}(\mathbf{R},\mathbf{k},t)\partial_{k_x}\rho_{\sigma_1\sigma'}(\mathbf{R},\mathbf{k},t) + \partial_{k_x}\rho_{\sigma\sigma_1}(\mathbf{R},\mathbf{k},t)\partial_x\Sigma_{\sigma_1\sigma'}(\mathbf{R},\mathbf{k},t)\Big]$$

$$+\frac{k_x}{m}\partial_x\rho_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t) + \frac{1}{4}\Big[\partial_x\big(h_x(\mathbf{k}) - i\sigma h_y(\mathbf{k})\big)\partial_x\rho_{-\sigma\sigma'}(\mathbf{R},\mathbf{k},t) + \partial_{k_x}\big(h_x(\mathbf{k}) + i\sigma' h_y(\mathbf{k})\big)\partial_x\rho_{\sigma-\sigma'}(\mathbf{R},\mathbf{k},t)\Big]$$

$$+\frac{1}{2}\sum_{\sigma_1}\Big[\partial_{k_x}\Sigma_{\sigma\sigma_1}(\mathbf{R},\mathbf{k},t)\partial_x\rho_{\sigma_1\sigma'}(\mathbf{R},\mathbf{k},t) + \partial_x\rho_{\sigma\sigma_1}(\mathbf{R},\mathbf{k},t)\partial_{k_x}\Sigma_{\sigma_1\sigma'}(\mathbf{R},\mathbf{k},t)\Big]. \qquad (15)$$

It is therefore noted that the corresponding coefficients of $\partial_x\rho_{\sigma\sigma'}$, $\partial_x\rho_{-\sigma\sigma'}$ and $\partial_x\rho_{\sigma-\sigma'}$ in the Bloch eqautions are

$$\frac{k_x}{m^*} + \frac{1}{2}\partial_{k_x}[\Sigma_{\sigma\sigma}(\mathbf{R},\mathbf{k},t) + \Sigma_{\sigma'\sigma'}(\mathbf{R},\mathbf{k},t)], \qquad (16)$$

$$\frac{1}{2}\partial_{k_x}\big\{[h_x(\mathbf{k}) - i\sigma h_y(\mathbf{k})]/2 + \Sigma_{\sigma-\sigma}(\mathbf{R},\mathbf{k},t)\big\}, \qquad (17)$$

$$\frac{1}{2}\partial_{k_x}\big\{[h_x(\mathbf{k}) + i\sigma' h_y(\mathbf{k})]/2 + \Sigma_{-\sigma'\sigma'}(\mathbf{R},\mathbf{k},t)\big\}, \qquad (18)$$

respectively. They are all k-dependent. Hence, similar to the simplified model, the interference effect is also important in the full kinetic equation. The kinetic equa-

tions (1) and the Poisson equation (3), together with the boundary conditions (9) and (10) can be solved numerically in an iterative manner to achieve the stationary solution.[30,31] The numerical results for a typical QW with width $a = 7.5$ nm, boundary spin polarization $N_{1/2}(0) = 2.05 \times 10^{11}$ cm^{-2} and $N_{-1/2}(0) = 1.95 \times 10^{11}$ cm^{-2} at temperature $T = 200$K are plotted in Fig. 2. In this computation, we only take into account the scattering due to longitude optical (LO) phonon. It can be seen from the figure that the surplus of the spin up electrons decreases rapidly along the diffusion direction, similar to the simplified model shown above.
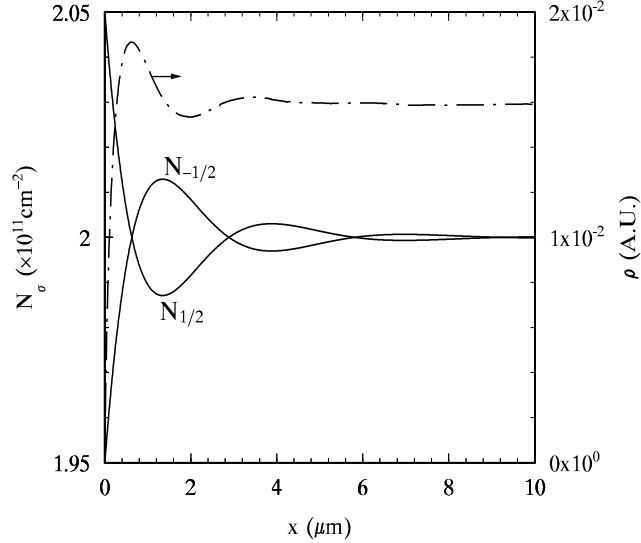
FIG. 1. Electron densities of up spin and down spin (solid curves) and incoherently summed spin coherence ρ (dashed curve) versus the diffusion length x. $B = 1$ T. Note the scale of the spin coherence is on the right side of the figure.
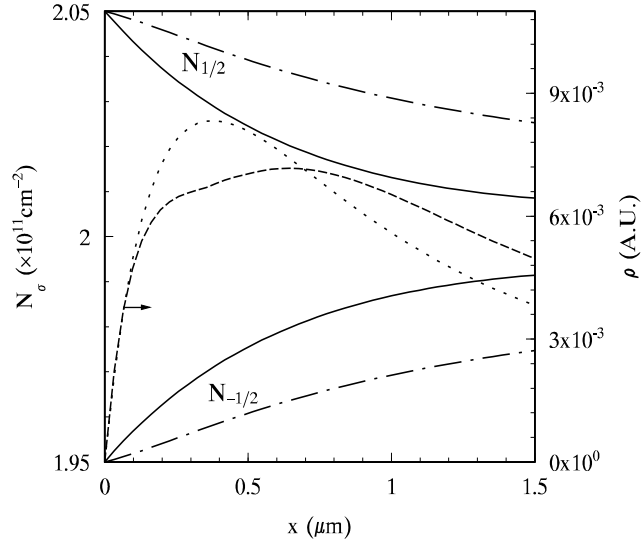

FIG. 2. Electron densities of up spin and down spin and the incoherently summed spin coherence versus the diffusion length x. Solid curves and dashed curve: N_σ and ρ from the full Bloch equations; Dash-dotted curves and dotted curve: N_σ and ρ from the equations without the interference effect. Note the scale of the spin coherence is on the right side of the figure.

The fast decay above is understood mainly generated by the decoherence from the interference effect due to the inhomogeneous broadening. Other dephasing effects such as those caused by the DP terms in Eqs. (5) and (6) as well as the spin conserving LO phonon scattering also

contribute to the decay. Besides, we pointed out that the inhomogeneous broadening effect combined with spin-conserving scattering can also cause spin dephasing.[19] Therefore, the above mentioned inhomogeneous broadening may also cause spin dephasing in the presence of LO phonon scattering. To compare the decoherence due to interference and the dephasing due to the DP term together with the scattering, we remove the interference effect in the transport equations by replacing k in the coefficients [Eqs. (16)-(18)] with $k = k_F$. Here k_F represents the Fermi wavevector. Therefore, if there is any decay of spin polarization along the diffusion direction, it comes from the spin dephasing. The numerical result is plotted in Fig. 2. It is shown clearly that the decay of spin polarization due to the dephasing effect alone (dash-dotted curves) is much slower than that due to the decoherence (interference) effect. In the figure we also plot the corresponding incoherently summed spin coherences ρ. One can see from the figure that both coherences ρ decay slowly and their decay rates are comparable when $x > 1$ μm. This further justifies what mentioned above that the fast decay of the spin polarization is mainly due to the interference effect.

In conclusion, we have set up many-body kinetic Bloch equations with spacial inhomogeneity. We reexamined the wildly adopted quasi-independent electron model and pointed out an important many-body spin decoherence effect which is missing in the single electron model. The new decoherence effect is based on an interference effect along the diffusion direction in spin transport due to the so called inhomogeneous broadening effect. We have shown that this inhomogeneous broadening effect can alone cause spin decoherence, even without the scattering and that the resulting decoherence is more important than the dephasing effect due to both the DP term and the scattering. Our study shows the inadequacy of the quasi-independent electron model. Therefore, it is important to use the full many-body theory to study the spin transport.

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